

REMARKS

Claims 1, 3 and 5 are pending in the application. Claims 1, 3 and 5 have been amended. Support for the amendments appear on pages 11-13 of the present specification. Claim 7 has been added to the application. Support for this claim appear in the paragraph bridging pages 8 and 9 and on pages 11-13 showing the exemplification of the present invention. No new matter has been added. Some formal typographical errors have been corrected in the specification, in particular, on pages 3 and 7 thereof as suggested in the Office Action.

Applicants gratefully acknowledge the indication that the rejection of claims 1 and 2 under 35 U.S.C. §102(b) in view of Wadlinger et al. has been withdrawn.

Claim 1 has been rejected under 35 U.S.C. §103(a) as being unpatentable over Wadlinger et al. Claims 3 and 5 remain rejected under 35 U.S.C. §103(a) as being unpatentable over Grasselli et al. Applicants traverse these rejections. They are similarly deficient.

Wadlinger et al. (Wadlinger) teaches in column 4, lines 60-65 that its new zeolite referred to as zeolite beta is different from any other known crystalline aluminosilicate in several ways. It continues to explain that statement in the following paragraph. It refers to the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio as being extremely high and extremely variable.

Wadlinger teaches the particular $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 10 to 200 relative to the composition of the reaction mixture used to form the zeolite beta, not to the zeolite beta itself. See col. 3, lines 64-67. Also note that the claims directed to the zeolite do not recite an $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio, whereas the claims directed to methods for making the zeolite do recite various $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratios.

Regarding the $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of the zeolite beta, Wadlinger states at col. 4, lines 73-75 that:

the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio is extremely high and extremely variable,
going from 10 to 100 and possibly as high as 150.

However, the “zeolite beta” in Wadlinger is very broad. For example, Wadlinger teaches that the compositions contemplated therein include not only the sodium form of zeolite beta but also “crystalline materials obtained from such a zeolite by partial or complete replacement of the sodium ion with other cations” (see col. 5, lines 38-44). Wadlinger teaches that:

[t]he sodium cations can be replaced, at least in part, by other ions including those of metals below sodium in the electromotive series, calcium, ammonium, hydrogen and combinations of such ions with one another. Particularly preferred for such purpose are the alkaline earth metal ions; Group 2 metal ions, ions of the transition metals such as manganese and nickel, the rare earth metals such as cerium, lanthanum, praseodymium, neodymium, samarium and mixtures thereof with each other and the other rare earth; and other ions, for example, hydrogen and ammonium which behave in zeolite beta as metals in that they can replace metal ions without causing any appreciable change in the basic structure of the zeolite crystal. The transition metals are those whose atomic numbers are from 21 to 28, from 39 to 46 and from 72 to 78 inclusive, namely scandium, titanium, vanadium, chromium, manganese, iron, cobalt, nickel, yttrium, zirconium, niobium, molybdenum, ruthenium, rhodium, palladium, platinum, hafnium, tantalum, tungsten, rhenium, osmium and iridium [emphasis added] (col. 5, lines 44-63).

Applicants submit that Wadlinger does not teach a particular $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio range for the hydrogen form of the zeolite beta. As noted previously, Wadlinger teaches that the $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of the zeolite beta is “extremely variable” (col. 4, lines 73-74).

To arrive at Applicants’ claimed catalyst, one skilled in the art reviewing Wadlinger would have to select a specific cation, i.e., hydrogen, from among those listed at col. 5, lines 44-63, to replace sodium ions in the zeolite beta and then select the specific $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of from 20-70 from $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratios that are “extremely variable, going from 10 to 100 and possibly as high as 150” (col. 4, lines 73-75).

Applicants respectfully submit that Wadlinger does not make obvious Applicants’ claimed catalyst because the reference does not specifically disclose the combination of a proton type β zeolite and an $\text{SiO}_2:\text{Al}_2\text{O}_3$ molar ratio of from 20 to 70, and that one skilled in the art would have to pick and choose among a long list of cations and a broad range of $\text{SiO}_2:\text{Al}_2\text{O}_3$ molar ratios to arrive at the claimed catalyst.

Applicants further point out that the claims now emphasize that the present invention is directed to a catalyst and a method that has substantial denitrification performance as well as durability. Further, these unexpected combined activities are set forth in the embodiments beginning at page 8 of the present specification. Also, claim 7 recites that the catalyst is grained to a particle size of 350-500 μm . Also, that the excessive oxygen referred to in the

independent claims include sulfur oxides. The claims further recite that this denitrification performance and durability occur even at temperatures of 300-400°C.

These results are shown in Table 1 in which catalyst 1 and 2 are according to the invention and show significant removal efficiency compared to catalyst that are not according to the invention for the reasons indicated in the exemplification of samples 4 -11. Further, Applicants point out that sample 6 drawn to catalysts C3 includes a ZMS5 as opposed to a zeolite beta.

Table 1

Sample	Catalyst	Reducing agent	NOx removal efficiency (%)		
			300.°C	350.°C	400.°C
1	Catalyst 1	Methanol	64	88	95
2	Catalyst 2	Methanol	59	81	90
3	Catalyst 1	Dimethyl Ether	64	84	93
4*	Catalyst C1	Methanol	18	27	35
5*	Catalyst C2	Methanol	22	46	56
6*	Catalyst C3	Methanol	17	32	51
7*	Catalyst C4	Methanol	42	66	67
8*	Catalyst C2	Dimethyl Ether	24	44	58
9*	Catalyst C3	Dimethyl Ether	15	35	52
10*	Catalyst 1	Propylene (500ppm)	27	40	61
11*	Catalyst 1	Propylene (1,000ppm)	29	45	67

Further, these significant results relating to catalyst activity are shown in Table 2.

Table 2

Sample	Catalyst	Reducing agent	NOx removal efficiency (%)	
			Before durability test	After durability test
12	Catalyst 1	Methanol	88	83
13	Catalyst 1	Dimethyl Ether	84	80
14*	Catalyst C2	Methanol	46	41
15*	Catalyst C3	Methanol	32	30
16*	Catalyst C4	Methanol	66	54
17*	Catalyst C3	Dimethyl Ether	35	32

Again, catalyst 1 is utilized in samples 12 and 13 and show significant improvement and durability compared to catalyst not according to the present invention, which are samples 14-17.

The substantial increases over the comparative examples including a ZSM5 catalyst are significant advances in the art. This is not taught in the applied reference.

Grasselli et al. (Grasselli) teach in column 4, zeolites including ZSM5 and in the latter part of the paragraph at the end of column 4 stating that zeolite beta is also included having a specific constraint index. This teaching of combining all of these zeolites is carried through in dependent claim appearing on column 12 of the reference.

The catalyst disclosed in Grasselli comprises a film of interconnected zeolite crystals bonded to a substrate surface (see, e.g., col. 2, lines 31-32, and col. 3, lines 21 – col. 4, line 2)). Grasselli teaches that:

Advantageously, in the catalyst structure for use herein the zeolite film is strongly bonded to the surface of a substrate so that the mechanical integrity of the film is maintained when the structure is exposed to high flow rates of gases or liquids. (col. 3, lines 3-7).

In the manufacture of the catalyst structure for use herein, a zeolite film is synthesized as bonded to a substrate. “Bonded” is intended to mean that the film is strongly adherent to the surface of a substrate and remains substantially adherent when subjected to conditions of catalysis, particularly high flow-through of gases and liquids. (col. 3, lines 28-34).

Thus, in Grasselli, the presence of the substrate is an important feature of the zeolite catalyst therein.

Regarding the zeolite catalysts produced by the method disclosed therein, Grasselli teaches that “[t]ypical zeolites to be synthesized according to this method are characterized by a Constraint Index of about 1 to about 12” (col. 4, lines 34-36). Regarding the $\text{SiO}_2\text{:Al}_2\text{O}_3$ molar ratios for the zeolites produced therein, Grasselli teaches that:

These zeolites may be produced with differing silica:alumina ratios ranging from 12:1 *upwards*. They may, in fact, be produced from reaction mixtures from which aluminum is intentionally excluded, so as to produce materials having extremely high silica:alumina ratios which, in theory at least may extend *up to infinity*. Silica:alumina ratios of *at least*

30:1 and higher will be common for these zeolites, e.g., 70:1, 200:1, 500:1, 1600:1 or even higher (col. 4, lines 52-61).
[emphasis added]

However, the Grasselli teachings cited above only relate to ZSM-type zeolites, not to proton type β zeolite catalysts. Regarding Zeolite Beta, Grasselli teaches that this zeolite has “a Constraint Index in the range of approximately 0.6-2.0” (col. 4, lines 65-67). However, this teaching does not relate to the $\text{SiO}_2:\text{Al}_2\text{O}_3$ molar ratio of the Zeolite Beta. Grasselli does not teach any correlation between the Constraint Index of a zeolite and its $\text{SiO}_2:\text{Al}_2\text{O}_3$ molar ratio. Thus, for at least this reason, Applicants respectfully submit that Grasselli does not teach or suggest a proton type β zeolite catalyst having an $\text{SiO}_2:\text{Al}_2\text{O}_3$ molar ratio of 20-70 or its use in purifying exhaust gas.

Grasselli’s teachings are specific and cannot be ignored in any combination with Wadlinger. The resulting product would differ from that presently claimed. Certainly, the unexpected results are not taught in either the Wadlinger reference or Grasselli. Actually, Grasselli teaches that the results would at least be similar for the zeolites listed including zeolite beta which has different crystallinity and the catalyst ZSM5.

However, in the presently claimed invention, the results significantly differ from the use of a catalyst in sample 6 drawn to ZSM5. Similar results are taught for samples 15 and 16 in Table 2. The present teachings show that the different zeolites differ significantly in removal efficiency at low temperatures as well as for durability. With regard to ZSM5 and sample 6, at 300°C, the removal efficiency is 17 compared to samples 1-3 having an efficiency from 59-64%. See Table 1. In Table 2, the durability is shown for catalyst C3 of either 32 or 35% whereas for catalyst according to the invention, they are over 80% in durability. Consequently, the combination of references or the references alone do not teach the presently claimed catalyst or method, particularly considering the broad disclosure of Wadlinger and the restrictive disclosure of Grasselli.

A reference which does not highlight a claimed mixture, among the many dozens disclosed, or suggest a claimed ratio, or where one skilled in the art would have to choose judiciously from a genus of possible combinations, would not likely render obvious claims drawn to the specific mixture or compound. In re Sivaramakrishnan, 213 USPQ 441 (CCPA 1982). Similarly, a generic formula which encompasses a vast number of compounds does not describe and thus anticipate or render obvious all compounds embraced therein merely because they are within the scope of the formula. In re Petering, 133 USPQ 275 (CCPA

1962). There can be no anticipation or obviousness where a reference is so broad that the likelihood of arriving at the claimed composition would be the same as discovering the combination of a safe by an inspection of its dials. Ex parte Garvey, 41 USPQ 583 (POBA 1939). Neither is anticipation or obviousness made out by a hindsight selection based on an Applicant's disclosure of variables of a broad generic disclosure. In re Rushig, 145 USPQ 274 (CCPA 1965).

The "non-preferred" as well as the "preferred" portion of a reference is pertinent from what it teaches to one skilled in the art. In re Meinhardt, 157 USPQ 270 (CCPA 1968). Similarly, teachings of a reference which would lead away from the claimed invention as well as those portions which would render aspects thereof as obvious are pertinent. In re Marshall, 198 USPQ 344 (CCPA 1978). It is not within the frame work of 35 U.S.C. §103 to pick and choose from the relevant prior art only as much as will support a holding of obviousness, to the exclusions of other parts necessary to the full appreciation of what the prior art suggests to one skilled in the art. In re Wesslau, 147 USPQ 391 (CCPA 1965).

Applicants respectfully refer the Examiner to the comments concerning the results as well as the combination of zeolites including ZSM5 and zeolite beta which teach away and overcome the subject rejections.

Catalysts are generally considered unpredictable merely from the chemical nature of the catalyst. The references considered alone or in combination do not show removal efficiency nor durability and in particular the comparative examples in the present specification establish an unpredictability for a specific zeolite beta. It further teaches a significant difference between the zeolite beta of the present invention and that of ZSM5. The references as pointed out teach in essence no such effects as taught by Applicants or teach equivalency for zeolites including both ZSM5 and zeolite beta. This establishes non-obviousness.

Further, Applicants point out that the present invention is directed to a granular catalysts which does not necessitate support rather than films. While Applicants' invention does not exclude being supported they do provide an option due to their characteristics. The prior art is restricted. Applicants' invention is not. The distinction among the claims through use of the phrase "consisting of" and the phrase "comprises" emphasizes a distinction that does not exist in the applied prior art relative to the comment set forth above concerning Grasselli.

For all the reasons advanced above, Applicants respectfully submit that the rejections of claims 1, 3 and 5 have been overcome and should be withdrawn.

For all the reasons advanced above, Applicants respectfully submit that the application is in condition for allowance and that action is earnestly solicited.

If any fees are due in connection with the filing of this Amendment, such as fees under 37 C.F.R. §§ 1.16 or 1.17, please charge the fees to our Deposit Account No. 02-4300; Order No. 034145.002.

Respectfully submitted,

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